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ABSORPTION OF ELECTROMAGNETIC RADIATION IN THE TRANSPARENT REGI--ETC(U)
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UNCLASSIFIED URI-9804/4528 ARO-12900.6-R-P NL

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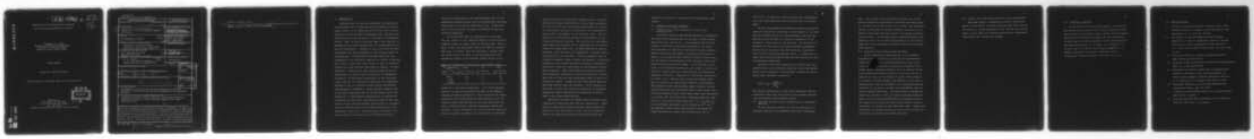

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ABSORPTION OF ELECTROMAGNETIC RADIATION IN THE TRANSPARENT REGIME OF SOLIDS

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FINAL REPORT

Grant No: DAHC04-75-G-0060

Period Covered: 14 October 1974 to 13 October 1975

Submitted to
Department of the Army
U. S. Army Research Office
P.O. Box 12211
Research Triangle Park, North Carolina 27709

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER

2. GOVT ACCESSION NO.

3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle)
Absorption of Electromagnetic Radiation in the Transparent Regime of Solids.

5. TYPE OF REPORT & PERIOD COVERED
Final Report.
10/14/74 - 10/13/75

6. PERFORMING ORG. REPORT NUMBER
14 Oct 74-13 Oct 75.

7. AUTHOR(s)
Shashanka S. Mitra

8. CONTRACT OR GRANT NUMBER(s)
DAHC04-75-G-0060 new
URI — 9804/4528

9. PERFORMING ORGANIZATION NAME AND ADDRESS
University of Rhode Island
Electrical Engineering Department
Kingston, Rhode Island 02881

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS

11. CONTROLLING OFFICE NAME AND ADDRESS
U. S. Army Research Office
Post Office Box 12211
Research Triangle Park, NC 27709

12. REPORT DATE
March, 1977

13. NUMBER OF PAGES
12p.

14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)
R. A. Shatas
U. S. Army Missile Command
Redston Arsenal, Alabama 35809

15. SECURITY CLASS. (of this report)
Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)
Approved for public release; distribution unlimited.

ARO 12900.6-R-P

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

ACCESSION for
NTIS White Section ☒
DDC Buff Section ☐
UNANNOUNCED ☐
JUSTIFICATION
BY
DISTRIBUTION/AVAILABILITY CODES
Dist. A.L. and/or SPECIAL
A

18. SUPPLEMENTARY NOTES
The findings in this report are not to be construed as an official Department of the Army position, unless so designated by other authorized documents.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)
transparent solids, laser radiation, multiphoton processes, anharmonic molecules, laser induced damage, direct gap semiconductors.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)
Interaction of high power laser radiation with matter has been considered. In particular, one- and multiphoton absorption coefficient in direct gap semiconductors have been calculated using a number of theoretical models and compared with experimental data. Selective multiple photon absorption by an anharmonic molecule has been treated including the effects of the rotational sublevels and the Stark broadening of the energy levels. The applicability of the extended avalanche breakdown processes to describe window surface damage by pulsed CO_2 → next page

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Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

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has been examined

Block 20 Abstract cont.

lasers. A low-loss isolator for 5.26 μm radiation utilizing the Zeaman effect in gases has been proposed.

I. INTRODUCTION

Although much work has been performed on measurements and interpretation of light absorption by opaque or nearly opaque solids, it is surprising to note that until recently relatively little reliable experimental data, and much less theoretical work was available on the nature of transparent solids. This, in spite of the fact that a vast majority of engineering and device applications of a solid depend on its optical transparency. Needless to say, all solids are both transparent and opaque depending on the spectral region of consideration. The absorption processes that limit the transparency of a solid are either due to lattice vibrations, as in ionic or partially ionic solids, or due to electronic transitions, both intrinsic and impurity induced. For most materials, a sufficiently wide spectral window exists between these two limits, where the material is transparent. In general, the absorption coefficient, in the long wavelength side of, but sufficiently away from the fundamental absorption edge, is relatively structureless and depends exponentially on frequency. Recent evidence suggests that in the short wavelength side of the one-phonon region, but beyond two- or three-phonon singularities, the absorption coefficient of both polar and nonpolar solids is also relatively structureless and depends exponentially on frequency. The main contributions to the residual absorption in the transparent regime are: (i) multi-phonon processes, (ii) various defects and impurities, (iii) phonon-assisted

electronic transitions in the long-wavelength tail of the fundamental absorption edge, and (iv) multiphoton electronic transitions in the case of high photon flux. In this project we have primarily focussed our attention on the last mentioned mechanism.

It is well know that the excitation of an electron from the valence to the conduction band of a solid requires a photon energy, at least, equal to the band gap. This, in fact, is only the predominant process by which an electron-hole pair is created. When the band gap is several times the photon energy, as shown in the following table, the one-photon process is not allowed, because energy is not

Order of Multiphoton Ionization for a Few Common Solids

Material / $\lambda(\mu m)$	GaAs $E_g \simeq 1.47$ eV	Si $E_g \simeq 0.8$ eV	KCℓ $E_g \simeq 8.2$ eV
10.6	13	7	71
1.06	2	1	8
0.694	1	1	5

conserved in the direct transition. It is still possible, however, to create an electron-hole pair in the material through multiphoton absorption. This process is not easily detectable with normal levels of light intensity, but can become a substantial fraction of the total absorption process, if high intensity beams are propagated through the material. Under illumination by intense coherent radiation sources presently available, many optoelectronic materials

thus are driven into a nonlinear response region. We have analyzed processes contributing to the nonlinear absorption of light by electronic transitions taking place as a result of a multiphoton process in a dielectric or a semiconductor solid usually transparent to low intensity light. Such absorption at high radiation fluxes is describable by a nonlinear differential equation. For the case of a weak radiation field, this equation can be linearized and its solution yields the Lambert-Beer law. At high intensities, however, nonlinear terms dominate, and consequently, a general analytic solution is not available. Physical arguments must be invoked to separate terms in powers of intensity. Electronic transition rates induced by photon processes of appropriate multiplicity enter as coefficients in the nonlinear differential equation. These coefficients have been calculated from fundamental properties of opto-electronically interesting solids at selected laser wavelengths using a variety of theoretical models. Comparison with experimental data has been made whenever they are available. As an extension of this work, we have also carefully considered the available theoretical models describing the fundamental absorption edge of direct gap semiconductors.

Research on a number of related topics involving laser light-matter interaction have also been carried out. These include, irreversible surface and bulk damage produced by high power laser radiation in solids, multiple photon absorption by anharmonic molecules and laser-induced dis-

sociation, and design of isolaters for selected laser wavelengths.

II. SUMMARY OF RESEARCH PERFORMED

2.1 Linear and Non-linear Absorption in Direct Gap Semiconductors

A critical review of multiphoton absorption coefficients for direct band semiconductors of interest in laser optical applications was carried out. The two-photon nonlinear absorption coefficients have been calculated in the second order perturbation employing interconduction band transition model of Braunstein and Ockman, two-valence band intraconduction or intravalence band transition model of Basov etal., and the Keldysh electromagnetic field "dressed" valence and conduction band wavefunction model. In all models, corrections to the original calculations were needed to account for effective masses, dielectric constants and the energy band dispersion relations. Comparison of theoretical predictions at 0.694, 1.06, 1.318 and 10.6 μm laser wavelengths shows that, in general, the Basov model slightly over-estimates, the Braunstein model substantially under-estimates the nonlinear absorption, and the Keldysh model yields second order nonlinear absorption about three times larger than Braunstein. It is shown that the second order nonlinear absorption coefficient in the photon energy range $E_g/2 < \hbar\omega < E_g$ where E_g is the forbidden gap width, can be as high as 10^{-5} cm W^{-1} and therefore under certain circumstances can exceed the linear absorption at laser flux intensities as low as

10^5 W cm^{-2}. In addition, nonlinear absorption coefficients up to the fifth order have been estimated from the Keldysh model.

We have also adapted the Keldysh multiphoton absorption theory to one-photon transitions in semiconductors. We find that the Keldysh theory is in good agreement with both the absolute values and with the frequency dependence of the absorption coefficients of InSb and GaAs. We have also re-examined the often used first-order perturbation approach and derived an expression for the absorption coefficient using the $\bar{k}\cdot\bar{p}$ method and parabolic bands. A comparison between theory and experimental data has been carried out without adjustable parameters.

One-photon absorption coefficients of direct gap semiconductors are next calculated using the first order perturbation theory assuming a two band model in which the interband energy difference is given by

$$E(k) = E_g\left(1 + \frac{\hbar^2 k^2}{m^* E_g}\right)^{1/2}$$

The results obtained are in much better agreement with the experimental data than the results of first order perturbation calculations using parabolic bands.

2.2 Selective Multiple Photon Absorption by an Anharmonic Molecule

We have discussed a model of selective absorption of infrared radiation by an anharmonic molecular vibrational

mode. The effects of the rotational sublevels and of the energy dissipation mechanisms are described phenomenologically. We show that in the context of our model, the absorption process is described by coupled nonlinear equations for the complex vibrational amplitude and for the average excitation quantum number. Two distinct dynamic regimes (coherent and adiabatic limit) can be identified depending on the relative magnitude of the rotational relaxation time and of the laser pulse duration.

2.3 Pulsed CO_2 Laser Window Damage Processes

We have examined the applicability of the extended avalanche breakdown process to describe the semiconductor window surface damage by pulsed high pressure CO_2 lasers. In the extension to optical frequencies of Shockley's avalanche theory construed originally for a dc electric field, a hard momentum-reversing collision of the free carrier is required to occur at the instant of optical field reversal. In contrast to alkali halides, the mobility is very high in semiconductors, and the probability of a momentum-reversing collision is too small to account for experimental observations of damage thresholds. Alternate models of damage processes are discussed. They are based on quantum mechanical photon assisted tunneling probability calculations between the bound and the quasi-free states and resonant collective excitations of carriers in the quasi-free state. Theoretical predictions for maximum pulsed CO_2 laser fluxes are made for some typical semiconducting window materials.

2.4 Isolator for 5.26 Microns Based on a Low Loss Rotator

The Zeeman effect in gases may be used to rotate the plane of polarization of plane polarized light with low loss of intensity. Design parameters for an isolator for 5.26 μm based on this effect are presented which show a significant improvement over conventional designs.

III. SCIENTIFIC PERSONNEL

In the spirit of the post-LRCP program, the research described here was carried out in close collaboration with the personnel of the Quantum Physics Group, Physics Research Directorate, Army Missile Command, and other consultants. In particular, the following people closely participated in the reported research. H. C. Meyer, R. A. Shatas, J. D. Stattler and G. A. Tanton (Army Missile Command); L. M. Narducci (Worcester Polytechnic Institute); and A. Vaidyanathan (graduate student, University of R. I.).

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